

Epitaxially grown sputtered LaAlO_3 films

A. E. Lee, C. E. Platt, J. F. Burch, and R. W. Simon
TRW Space & Technology Group, Redondo Beach, California 90278

J. P. Goral and M. M. Al-Jassim
Solar Energy Research Institute, Golden, Colorado 80401

(Received 25 June 1990; accepted for publication 6 September 1990)

We have grown crystalline thin films of LaAlO_3 using off-axis rf sputtering from a single stoichiometric target. The films grow epitaxially on SrTiO_3 and LaAlO_3 (100) substrates as well as on $\text{YBa}_2\text{Cu}_3\text{O}_7$ thin films. We report on the growth conditions used to make these films, the properties of the films, and the properties of bilayer and trilayer structures containing both LaAlO_3 and $\text{YBa}_2\text{Cu}_3\text{O}_7$ films. Transmission electron microscopy cross-sectional and x-ray diffraction analyses indicate that all the constituent films in the multilayers grow epitaxially and that the interfaces between the films are sharply defined. Preliminary transport measurements on these multilayers show that LaAlO_3 can be used for dielectric layers in a variety of high-temperature superconductor electronic circuits.

Superconductive electronics as currently implemented is a multilayer technology incorporating thin films of superconductors, normal metals, and dielectrics. Dielectric films both isolate conducting layers from one another and form an integral part of critical device structures such as high-frequency transmission lines and Josephson junctions. In general, dielectric films used in superconductive electronics must be smooth, uniform in thickness, pinhole-free, and must have desirable high-frequency properties.¹ The task of developing such films for high-temperature superconductor (HTS) device technology is complicated by the unique characteristics of the superconducting cuprates (anisotropy, chemical reactivity, etc.) which impose additional requirements upon the dielectric films. In particular, the highly anisotropic properties of the HTS cuprates necessitate the use of lattice-matched substrate materials in order to promote the growth of highly oriented films.² To produce similarly ordered films in the upper layers of multilayer structures, intermediate films must transmit the epitaxy begun at the original superconductor-substrate interface. As a result of such considerations, the prime candidate materials for dielectric films for HTS multilayer technology turn out to be the substrate materials themselves.

As soon as we identified LaAlO_3 as a desirable substrate for HTS films,^{3,4} we also realized its potential as a compatible thin-film dielectric. The development of the off-axis sputtering technique for oxide films⁵ provided a promising technique for depositing LaAlO_3 films. Since the superconductor-dielectric-superconductor trilayer is the basic structure for both microwave transmission lines and sandwich-type Josephson junctions, we have concentrated on developing these trilayers using $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) and LaAlO_3 sputtered films. This letter reports our results in applying off-axis sputtering to the development of LaAlO_3 films and HTS multilayer structures.

The sputtering system used in this study is similar to the off-axis single target system which has gained wide use for deposition of HTS films. As shown in Fig. 1, our system contains two separately shuttered sputter guns mounted in the off-axis configuration. Both the LaAlO_3 and YBCO (or, in some cases, $\text{ErBa}_2\text{Cu}_3\text{O}_7$) sputter targets are sto-

ichiometric single-phase material and are epoxied onto copper backing plates to facilitate heat dissipation during sputtering. We have used both sintered powder⁶ and molten-processed sputter targets for the LaAlO_3 films with comparable results.

To produce trilayers, both the initial YBCO layer and the LaAlO_3 are deposited at a pressure of 70 mTorr (30 mTorr O_2 /40 mTorr Ar). The substrates are attached to a stainless-steel block using silver paste and are heated by low-voltage quartz lamps mounted in a gold-coated copper reflector. Growth temperature is monitored with a K-type thermocouple embedded in the substrate block and is held at 760 °C for the first two layers. The deposition rate of the LaAlO_3 is about 40 nm/h. Using the same conditions for the third layer as for the first results in reduced transition temperatures. Substantially raising substrate temperatures either destroys the superconducting state of the bottom layer or produces a rough flaky surface. We eventually found that using higher pressure (40 mTorr O_2 /120 mTorr Ar) and lower substrate temperature (700 °C) produces a top layer with good electrical properties and a smooth surface, and does not degrade the properties of the bottom layer.

The primary substrate material for this study was (100) oriented LaAlO_3 . For x-ray diffraction studies, we

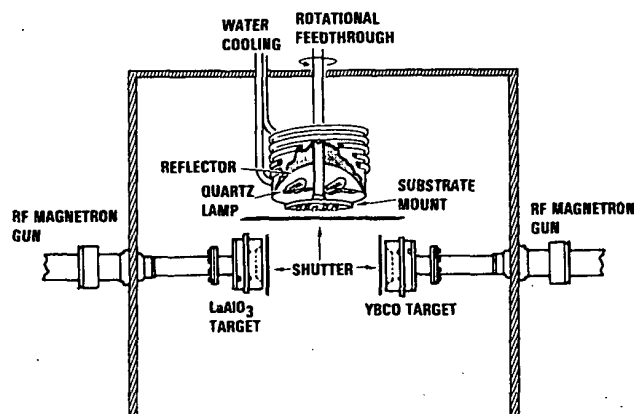


FIG. 1 Diagram of multisource off-axis sputtering system.

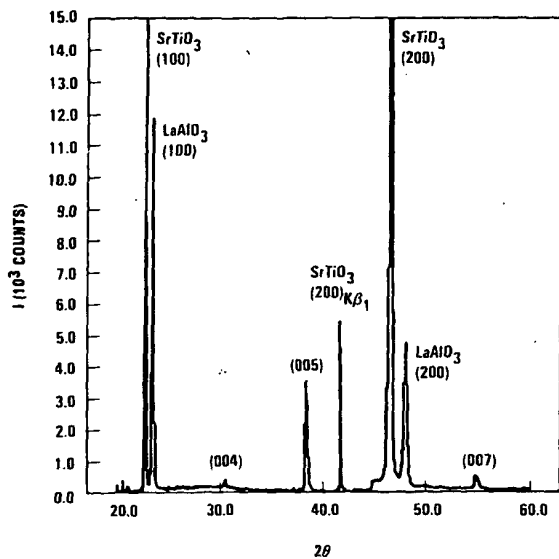


FIG. 2 XRD spectrum for LaAlO₃-on-YBCO bilayer grown on SrTiO₃ substrate.

used (100) SrTiO₃ substrates in order to distinguish peaks due to the films from those of the substrate. For this work we index the LaAlO₃ crystal (which is, strictly speaking, rhombohedral⁷) as though it were cubic.

Initially, we attempted to deposit LaAlO₃ on a variety of substrates (SrTiO₃, LaAlO₃, Si, YSZ, Al₂O₃ and MgO) under diverse temperature and pressure conditions. We found that only SrTiO₃ and LaAlO₃ substrates yielded crystalline-phase LaAlO₃ films; the other substrates resulted in amorphous samples. We obtained epitaxial growth of LaAlO₃ (100 orientation) using the (100) face of SrTiO₃, and the resultant films were smooth and featureless in scanning electron microscopy (SEM) images down to the 100 nm length scale.

These results emphasize the importance of choosing a lattice-matched substrate for epitaxial growth of LaAlO₃ at relatively low temperatures. Furthermore, in order to grow featureless LaAlO₃ films, it was necessary to use substrates with extremely good surfaces. Thus it was not surprising to find that LaAlO₃ films can only be deposited with smooth and continuous coverage on HTS films when the HTS films themselves are smooth.

To produce YBCO-LaAlO₃ bilayers, we first deposited 200-300 nm of YBCO on LaAlO₃ or SrTiO₃ substrates and then deposited 40-200 nm of LaAlO₃ under the conditions described above. A typical x-ray diffraction pattern of such a bilayer is shown in Fig. 2, which indicates that the orientation of the YBCO and the LaAlO₃ are (001) and (h00), respectively. No impurity lines were detected. The (005) peak width (FWHM) full width at half maximum is about or less than 0.2° in the typical YBCO film, and the rocking curve data shows the width of 0.16°. The peak width of the (100) LaAlO₃ is 0.15°, which is similar to that of adjacent YBCO peak (003). Diffraction patterns are obtained from samples on SrTiO₃ substrates in order to distinguish the LaAlO₃ thin-film lines.

By interrupting the trilayer process at various stages for measurements, we were able to study the effects of

multiple depositions on film morphology and on the superconducting transition temperature (T_c) of the bottom layer. We used an inductive technique to measure the transition temperature of the dielectric-covered YBCO film in a bilayer. We found that the transition was essentially unaffected by the presence of the LaAlO₃ film.

We also used the YBCO/LaAlO₃ bilayers to investigate the insulating properties of the dielectric films. This was accomplished by argon ion beam etching the LaAlO₃ layer to expose some of the YBCO base layer, depositing 100 μ m silver contact pads all over the wafer, and measuring the continuity between silver pads located on and off the dielectric layer. Out of a random selection of 15 Ag contact pads, 12 registered open circuits across the 200 nm LaAlO₃ film and 3 were completely shorted. The shorted contacts were subsequently seen to have large cracks in SEM micrographs.

In subsequent experiments, we typically observed room-temperature resistances above 100 k Ω for similar 100 μ m contact pads on top of 100 nm LaAlO₃ layers. These resistances increase rapidly upon cooling, indicating non-metallic shorting between the layers. Such results, while not ideal, are acceptable for many applications. We are currently investigating the rf dielectric properties of these films. We have not investigated the properties of ultrathin LaAlO₃ layers such as would be found in a tunnel junction structure.

The structure of greatest interest is the trilayer containing two YBCO films. Our results on such trilayers indicate that both YBCO layers are *c*-axis oriented and sharp LaAlO₃ x-ray peaks (seen on SrTiO₃ substrates) demonstrate that the dielectric layer has maintained its crystalline order.

We examined interfacial areas between the layers and the substrate of these trilayers using transmission electron microscopy (TEM) cross sectioning. All three layers show sizable regions of single-crystal, defect-free, epitaxial growth with the YBCO *c*-axis perpendicular to the substrate. Figures 3(a) and 3(b) demonstrate that interfacial transitions between YBCO and LaAlO₃ can be continuous in atomic stacking while exhibiting an abrupt change in atomic species. Figure 4 shows the selected-area diffraction pattern of the top YBCO layer of a trilayer sample; the electron beam is perpendicular to the film surface. The single-crystal-like diffraction pattern over a several square-micron area is characteristic of a high quality epitaxial YBCO film.

We used a combination of $R(T)$ and $M(T)$ measurements to study the superconducting transitions at various stages of trilayer deposition. It should be noted that inductive techniques are of limited use for evaluating the properties of structures containing multiple superconducting layers since the highest T_c layer will effectively screen out the other layers. Film patterning was required to look at the completed trilayers. We note that the YBCO films used for these studies were optimized for smooth surface morphology rather than for high T_c . As YBCO film processing now stands, 90 + K films are distinctly rougher than lower T_c specimens. Thus, the T_c of the bottom YBCO layer was

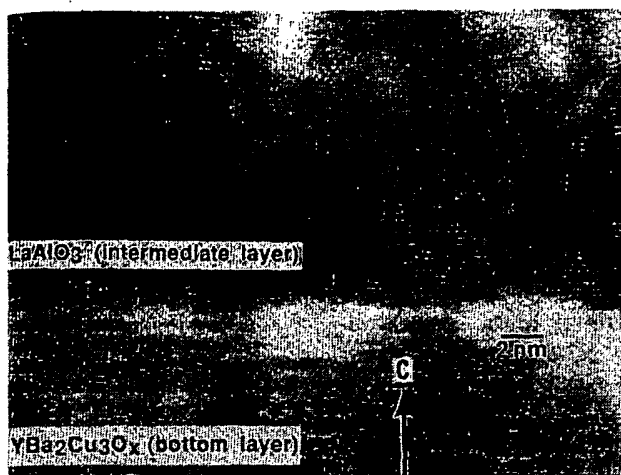
LaAl
YBa
(a)

YF
(b)

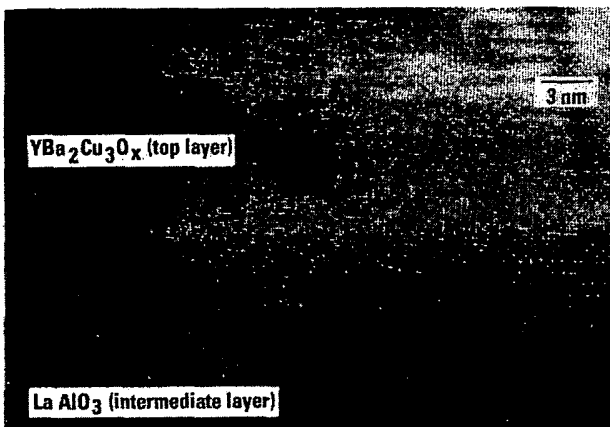
FIG
YBC
of in

typi
 T_c
high
ma;

FIG
tril:



(a)



(b)

FIG. 3. (a) Cross-sectional TEM image of interface between bottom YBCO layer and LaAlO_3 film in trilayer. (b) Cross-sectional TEM image of interface between top YBCO layer and LaAlO_3 film in trilayer.

typically 82–87 K, although it was not uncommon for the T_c of the top film to be higher, perhaps because of the higher sputtering pressure for this layer.

We have successfully used the off-axis single target magnetron sputtering technique to produce crystalline

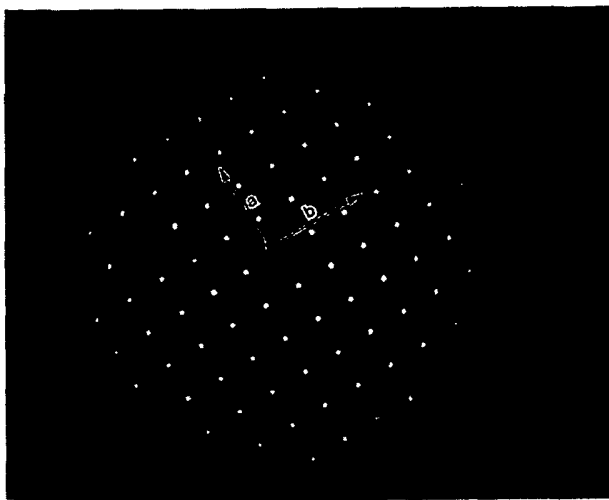


FIG. 4. Selected-area diffraction pattern in TEM of YBCO layer in trilayer. The electron beam is perpendicular to the film surface.

films of LaAlO_3 on lattice-matched substrates using the identical deposition procedure developed for $\text{YBa}_2\text{Cu}_3\text{O}_7$ films and have incorporated these films into multilayers containing $\text{YBa}_2\text{Cu}_3\text{O}_7$ or $\text{ErBa}_2\text{Cu}_3\text{O}_7$ films also made by off-axis sputtering. The YBCO and LaAlO_3 layers of trilayer structures are highly oriented, smooth, and of good crystalline quality. We see no evidence of interdiffusion between LaAlO_3 and YBCO films and the LaAlO_3 films serve as a nearly ideal template for epitaxial growth of YBCO.

The initial use for the trilayers developed here will be for passive microwave components, transmission lines, and coupling coils for superconducting quantum interference devices. Simple crossover structures like the coils are an important element in multilayer device technology that should be easily fabricated using these films. Similar work has already been done using SrTiO_3 laser evaporated films.⁸ The low dielectric losses in LaAlO_3 and its relatively low dielectric constant⁹ of ~ 26 (a higher value than we originally reported) make the microwave applications look particularly promising.

Of course, the most significant application for dielectric films in superconductive electronics is their use as tunneling barriers. The materials' demands on both the superconductor films and the dielectric layers are greatest for this application, but at least in principle, thin films of LaAlO_3 may be an excellent choice for this purpose as well. At this stage of technology development, LaAlO_3 appears to have all the desirable properties of a thin-film dielectric material compatible with the fabrication and processing of high-temperature superconductor circuits.

We gratefully acknowledge the collaborative efforts of C. Poirot and Professor B. Yazar at Colorado School of Mines in the synthesis of LaAlO_3 sputter target material and for rocking curve analysis. We also thank D. Harris at Charles Evans and Associates and L. Keller at Camet Research for their efforts in thin-film analysis. Finally, we express our gratitude to our colleagues K. P. Daly, M. S. Wire, and T. P. Neal at TRW for their contributions to this work. This research was funded by Office of Naval Research/Defense Advanced Research Projects Agency under contract N00014-88-C-0747.

¹See, for example, R. K. Hoffmann, *Handbook of Microwave Integrated Circuits* (Artech House, Norwood, MA, 1987), pp. 21–22.

²P. Chaudhari, R. H. Koch, R. B. Laibowitz, T. R. McGuire, and R. J. Gambino, *Phys. Rev. Lett.* **58**, 2684 (1987).

³R. W. Simon, C. E. Platt, A. E. Lee, G. S. Lee, K. P. Daly, M. S. Wire, J. A. Luine, and M. Urbanik, *Appl. Phys. Lett.* **53**, 2677 (1988).

⁴R. W. Simon, A. E. Lee, C. E. Platt, K. P. Daly, J. A. Luine, C. B. Eom, P. A. Rosenthal, X. D. Wu, and T. Venkatesan, *Science and Technology of Thin Film Superconductors*, edited by R. D. McConnell and S. A. Wolf (Plenum, New York, 1989), p. 337.

⁵C. B. Eom, J. Z. Sun, K. Yamamoto, A. F. Marshall, K. E. Luther, T. H. Geballe, and S. S. Laderman, *Appl. Phys. Lett.* **55**, 595 (1989).

⁶Synthesized by C. Poirot and B. Yazar at Colorado School of Mines, Golden, Colorado.

⁷S. Geller and V. B. Bala, *Acta Cryst.* **9**, 1019 (1956).

⁸J. J. Kingston, F. C. Wellstood, P. Lerch, A. H. Miklich, and J. Clarke, *Appl. Phys. Lett.* **56**, 189 (1990).

⁹M. C. Nuss, P. M. Mankiewich, R. E. Howard, B. L. Straughn, T. E. Harvey, C. D. Brandle, G. W. Berkstresser, K. W. Goossen, and P. R. Smith, *Appl. Phys. Lett.* **54**, 2265 (1989).